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New quantum properties of phonons and their detection

M. Artoni
Photonics, NASA/Goddard Space Flight Center, Greenbelt, MD 20771, U.S.A.

Joseph. L. Birman
Physics Department, The City College of New York, Convent Avenue at 138th Street New York, N.Y. 10031, U.S.A.

Abstract

We present a theoretical investigation on new and interesting properties of the phonon polarization field in solids. In particular, non-classical aspects of the phonon population and an experimental scheme that would enable one to detect them will be discussed.

1. *Introduction*

In recent years much interest has been devoted to the investigation of quantum effects that have no classical analogs, of which optical squeezing is the most ubiquitous one [1]. In view of the successful generation and detection of squeezed states of the electromagnetic (e.m.) field it is natural to ask whether analogous states exist and can be observed for other boson fields. Condensed matter exhibits a variety of bosons that, via the interaction with an external field, can be excited into a squeezed state in much the same way as done with photons.

It was previously pointed out [2] that in a phonon-polariton [3,4], a mixed mode in which an optical phonon is coupled to a photon, the photon component exhibits non-poissonian quantum statistics and optical squeezing [2,5]. Owing to the quadratic nature of the transformation that takes one from coupled bare phonons plus bare photons to a polariton, where phonons and photons appear as exactly *dual* particles, we demonstrate that the phonon component of a polariton exhibits analogous properties. We further analyse an experiment that would enable one to detect such delicate features in solids with an appreciable effect.

2. *Phonon-polariton*

Many topics in solids combine wave and particle aspects. Exactly as the photon describes the particle nature of the e.m. field, the phonon describes the particle nature of a lattice vibration [6]. Under certain conditions these two excitations may interact: at resonance, transverse optical phonons and photons couple and the character of the propagation inside the crystal is entirely changed. The pioneering work of Pekar [7], Fano [4], and Hopfield [8] has shown that eigenstates of the coupled system of a lattice vibration and radiation are composite *particles* made of photons and phonons, i.e. *phonon-polaritons* (polaritons hereafter). This represents the quantomechanical equivalent of the classical work of Huang [9] who first derived the dispersion for infrared active optical lattice vibrations of a cubic ionic crystal, showing that the actual modes propagating in the crystal are radiation-lattice coupled *waves*. A typical dispersion curve (no spatial dispersion) for a polariton is shown in Fig. 1 below.

3. *Squeezed states*

Squeezed states are quantum states in which the fluctuations in one of the phase quadratures of the field are reduced below the vacuum noise limit [1]. *Single* and *two*-modes squeezed states have

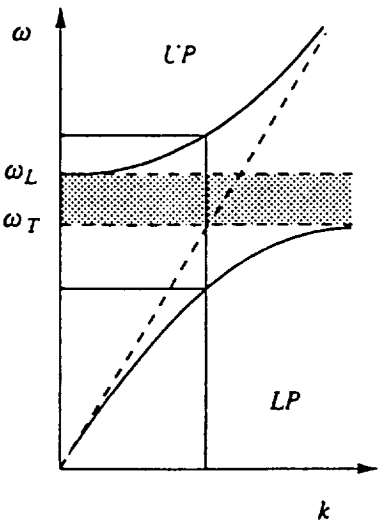

***Fig.1** Schematic polariton energy spectrum. UP and LP denote upper and lower dispersion branches. ω_T and ω_L are transverse and logitudinal phonon resonant frequencies.*

been extensively studied [1], and experiments [1] have already demonstrated their realizability in the case of an e.m. field.

If $\hat{a}_l$ and $\hat{b}_l$ denote two independent bosons, one can introduce [2] a two-mode squeezed state for the *mixed* boson $\hat{\gamma}_l = \alpha_l \hat{a}_l + \beta_l \hat{b}_l$ as a displaced state of the squeezed vacuum

$$|\gamma_{12}\rangle = \hat{D}(\gamma_{12}) \cdot \hat{S}^{\gamma}_{12}(r)|0\rangle = \\ = \{e^{-\frac{1}{2}(|\gamma_1|^2+|\gamma_2|^2)^2} e^{\gamma_1 \hat{\gamma}_1^\dagger} e^{\gamma_2 \hat{\gamma}_2^\dagger}\}\{e^{r(\hat{\gamma}_1\hat{\gamma}_2 - \hat{\gamma}_1^\dagger \hat{\gamma}_2^\dagger)}\}|0\rangle \tag{1}$$

$\hat{D}(\gamma_{12})$ and $\hat{S}^{\gamma}_{12}(r)$ are two-mode *displacement* and *squeeze* operators [10], respectively, whereas the other symbols have their usual meaning. r, the squeeze factor, mediates the coupling between the two modes 1 and 2. In the states $|\gamma_{12}\rangle$ these modes become so tightly correlated that they no longer fluctuate independently by even the small amount allowed in a coherent or vacuum state [1].

4. *Non-classical phonons*

The objective of this section is to show that a polariton state is a non-classical state and that the phonon counterpart associated with it exhibits non-classical features. We will restrict ourselves to *two-mode polariton coherent states*. In particular, these states are most suitable to describe the actual experimental scheme which we will discuss below. They can be constructed [2,11,12] from the polariton vacuum $|0\rangle_{pol}$, defined as $\hat{\eta}_{\pm k}|0\rangle_{pol} = 0$, $\hat{\eta}_{\pm k}$ being the polariton transformation [8]

$$|\eta_{\pm k}\rangle_{pol} = D(\hat{\eta}_k, \eta_k) D(\hat{\eta}_{-k}, \eta_{-k})|0\rangle_{pol}; \quad D(\hat{\eta}_{\pm k}, \eta_{\pm k}) \equiv e^{-|\eta_{\pm k}|^2/2}\, e^{\eta_{\pm k}\hat{\eta}^\dagger_{\pm k}} e^{-\eta^*_{\pm k}\hat{\eta}_{\pm k}} \tag{2}$$

$|\eta_{\pm k}|^2 = |\gamma_{\pm k} c_r + e^{2i\phi}\gamma^*_{\mp k} s_r|^2$ yields the average number of polaritons in the state (2), where the $\gamma_{\pm k}$'s are the eigenvalues of the bose operator $\hat{\gamma}_{\pm k} = \alpha_k^\pm \hat{a}_{\pm k} + e^{i\chi_k^\pm} \beta_k^\pm \hat{b}_{\pm k}$. The $\hat{a}$'s and $\hat{b}$'s are respectively photon and phonon bare field annihilation operators. The real parameters $\{\alpha_k^\pm, \beta_k^\pm, \chi_k^\pm, \phi, r_k\}$ are *mode* and *material* dependent [2].

One should focus at this point on the structure of the polariton vacuum state. It has been the object of an exhaustive study not only by us [2,13], but also other workers [14] though within different contexts. In this rather interesting work a crucial common result emerged: the polariton vacuum $|0\rangle_{pol}$ is unitarily related to the bare particles vacuum $|0\rangle$ by a transformation of squeezing [1,10], that is

$$|0\rangle_{pol} = \hat{S}(\hat{\gamma}_\pm, r_k)|0\rangle = e^{r_k(\hat{\gamma}_k\hat{\gamma}_{-k} - \hat{\gamma}_k^\dagger \hat{\gamma}_{-k}^\dagger)}|0\rangle \tag{3}$$

r_k gives the amount of squeezing in the mode k. It is then clear from Eq.s (2) and (3) that a polariton coherent state is an instance of the two mode squeezed state defined in Eq. (1), 1 and 2 referring to counterpropagating wavevector modes.

It also follows from Eq.s (2) and (3) that for those k's for which $S \cong 1$ the state $|\eta_{\pm k}\rangle_{pol}$ reduces to a bare particle coherent state, obtained by displacing the bare vacuum $|0\rangle$; but for those k's for which $S \neq 1$, owing to purely quadratic terms in the photon and phonon creation and annihilation operators, $|\eta_{\pm k}\rangle_{pol}$ acquires a significantly more complicated structure. The non-classical character of a polariton state is clearly related to the parameter r_k. Owing to the wide breadth of values that r_k can take on [2,5,15] one presume to create a polariton state with strong enough non-classical character so as to produce a sensitive result in the detection process. To this extent we recall from the results reported in [15] that, for a GaSb crystal, r_k may vary across the polariton spectrum between values bigger than 1 to values that are even two orders of magnitude smaller.

Non-classical phonons in a polariton would be commonly characterised by a non-classical probability density distribution of the number of phonons in the polariton state [1,16]. In a polariton coherent state, unlike a polariton number state, no definite number of polaritons exists, but a well defined probability corresponds to each polariton number with a distribution of probabilities known to be *classical* [1,2,16]. Nonetheless, the distribution of its phonon component is in general *not classical*, with the most striking effects occurring where the squeezed structure of a polariton is most enhanced.

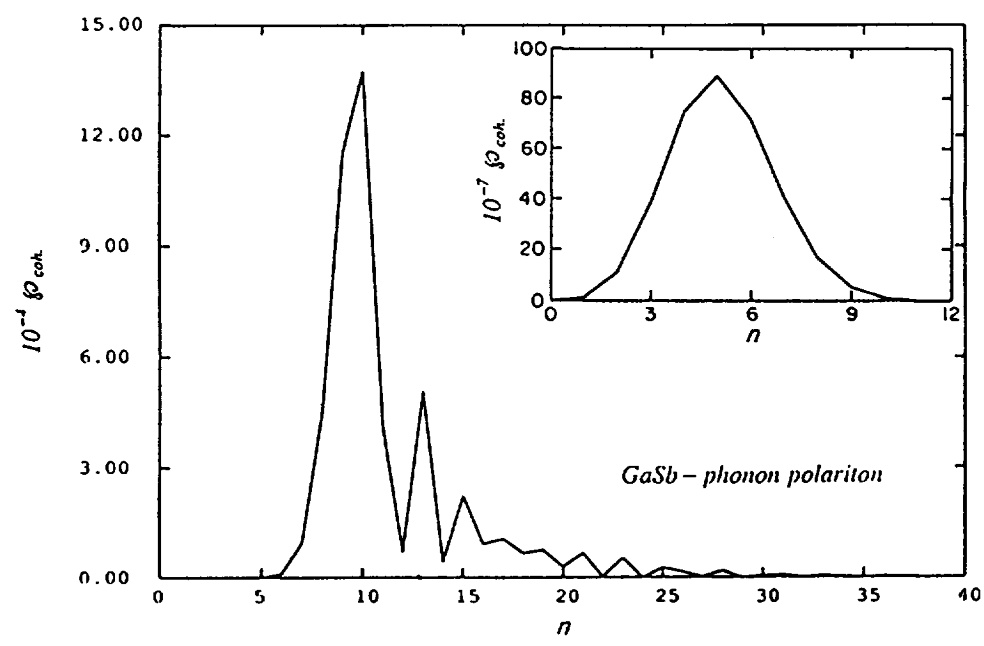

Fig.2 Probability distributions for observing n phonons in a GaSb-polariton [15] for the two modes $k_o = 10\,cm^{-1}$ $[r_o \approx 2.0]$ *(left) and* $k_o = 5.5\cdot 10^3\,cm^{-1}$ $[r_o \approx 5\cdot 10^{-2}]$ *(onset)*

The phonon number distribution in a polariton coherent state consists indeed of two contributions [15],

$$\wp_{coh} \equiv \wp_{coh}^{Poiss.}(n_{\pm k}) \cdot \wp_{coh}^{osc}(n_{\pm k}, r_k) \, , \tag{4}$$

a Poisson one ($\wp_{coh}^{Poiss.}$) whose structure remains as such through the whole spectrum, and an oscillating one ($\wp_{coh}^{osc}$) whose size depends on the polariton dispersion. For those modes k's for which $r_k \approx 0$ (Fig. 2 onset) $\wp_{coh}$ reduces to the Poisson distribution $\wp_{coh}^{Poiss.}$ (*classical limit*), whereas for those k's for which $r_k \neq 0$ (Fig. 2) the component $\wp_{coh}^{osc}$ will contribute with strong oscillations in the large n side of the distribution (*non-classical limit*). In this limit, quasi-periodic oscillations give rise to a remarkable effect of "quantization" in the phonon population which appears to be a distinctive non-classical feature of the phonon field in the polariton state (2). The two limits are illustrated in Fig. 2 through a numerical evaluation of Eq. (4) for GaSb polaritons [15].

5. Detection

The objective of this section is to address the question of how to probe the non-classical characteristics of the phonon field of a polariton discussed above. Probing the phonon number distribution associated with a specific structure of the polariton state is problematic if one decides to use particle-counting techniques analogous to those generally employed in the optical domain. *Conversely*, it would be possible to probe directly the non-classical structure of the polariton state that yields a non-classical phonon density distribution.

The idea consists in establishing coherence between two scattering processes that involve the absorption of two different wavevector phonon modes. Coherence would then produce constructive or destructive interference depending on whether the polariton is in a *non-classical* or *classical* state. Thus a measurement of the rate with which the probe scatters off of the phonon field of a polariton would provide a signature of the *non-classical* character of a polariton state.

This idea can be implemented as follows. Let a two-mode polariton be excited in a crystal, the modes referring to counterpropagating wavevectors of magnitude $|k_o|$ and frequency ω_o. Then let a two-components probe beam, having high coupling efficiency to the phonon part of the polariton (neutrons e.g.), impinge on the crystal: both probe components have incident energy ω_{inc}, but different wavevectors k_d and k_s (Fig. 3). The kinematics of the scattering process is described simply by the general conservation of energy and momentum (wavevector). Taking advantage of these laws, the input probe beam can be arranged so that the incoming probe is scattered into a given output state $|out\rangle_{pr}$ only when absorption off of the modes k_o and $-k_o$ occurs as schematically illustrated in Fig.3.

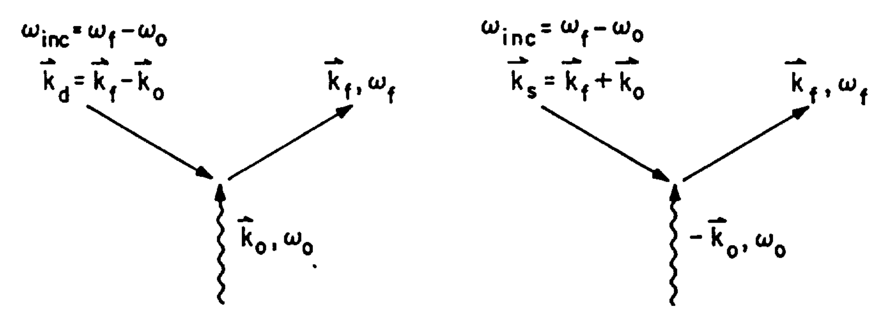

Fig.3 Scattering processes involving the absorption of two counterpropagating phonons with energy ω_o, and wavevectors k_o and $-k_o$. With the probe in the state $|in\rangle_{pr}$ (Eq. 5), the two processes can coherently interfere when the phonon field is squeezed.

Now let the detector be arranged so that only a probe in a final state of momentum $\boldsymbol{k}_f$ and energy ω_f be detected and let the incoming probe be in a coherent superposition of states with wavevectors $\boldsymbol{k}_d$ and $\boldsymbol{k}_s$,

$$|out\rangle_{pr} = \hat{c}^{\dagger}_{k_f} |0\rangle_{pr} \qquad |in\rangle_{pr} = [\, A\hat{c}^{\dagger}_{k_d} + e^{i\varphi_{pr}} B\hat{c}^{\dagger}_{k_s}]|0\rangle_{pr} \tag{5}$$

A, B and φ_{pr} are respectively the real amplitudes and relative phase of the two components probe. These parameters can be all made to be controllable.

The relevant scattering rate (lowest order), when a polariton is initially excited into a coherent state, is

$$\sigma^{pol}_{coh}(k_o, A, f^o, \varphi_{pr}) = \sigma^{Poiss.}(k_o, A, f^o, \varphi_{pr}) + s^2_{r_o}\sigma^{osc.}(k_o, A, f^o, \varphi_{pr}) \tag{6}$$

denoting by f^o the scattering amplitude for the process. The rate consists of two parts: one independent of $r_o \equiv r_{k_o}$ arising from the classical part of the distribution $\wp^{Poiss}_{coh}$ and one r_o-dependent coming from the oscillating counterpart $\wp^{osc}_{coh}$ (cf. Eq. 4). The relative size of these two contributions does play a significant role in determining the magnitude of the scattering rate. Namely, when polariton modes $|\boldsymbol{k}_o|$ are populated for which is $r_o \approx 0$ Eq. (6) is approximated by

$$\sigma^{pol}_{coh}(k_o, A, f^o) = \sigma^{Poiss.}(k_o, A, f^o, \varphi) \tag{7}$$

In this case we can show [15] that for suitable values of the amplitude A there exists a phase φ_{pr} for which $\sigma^{Poiss.} \to 0$ so that the lowest-order scattering can be completely inhibited. On the *contrary,* when polariton modes $|\boldsymbol{k}_o|$ are populated for which is $r_o \neq 0$ the second contribution ($\sim s^2_{r_o}$) in Eq. (6) is not negligible and can be shown to be always positive defined [15]. For these modes the rate turns out to be always greater or equal to $\sigma^{osc.}$, but never 0.

Hence rate measurements would permit one to discern the *non-classical* and *classical* character of a polariton coherent state. Destructive interference, able to suppress the rate with which a probe is scattered, is a signature of a classical polariton state, conversely constructive interference, yielding in principle a nonvanishing rate, is exhibited when the polariton state is non-classical (squeezed).

6. Discussion

Squeezed states, a familiy of pure quantum states having no classical analogue, have appeared in the literature since 1960's. In particular, extensive theoretical investigations for the realization of squeezed states of the electromagnetic field are also of long lasting. Only recently was the experimental realization of squeezed light with fewer quantum fluctuations than the vacuum achieved. To date there have been no reports, however, of the existence of non-classical states in condensed media, but the situation appears to be rather favorable for polaritons. This crystal mixed quasiparticle appears to be indeed a promising place where to look for non-classical states of light and other bosons, such as e.g. phonons, especially if extremely low loss crystals can be obtained.

A further extension of this work would include considerations on the physical origin of the non-classical effects discussed above. Intermode correlation resulting from the quadratic transformation that takes one from coupled bare photon plus bare phonon to polaritons is a plausible origin for the squeezed structure of a polariton state and ultimately for the non-classical features of the phonon component associated with it.

In a real material elementary excitations and quasi-particles normally experience dissipation and phase destroying processes that may degrade the structure of the non-classical state inside the medium. For a complete treatment and in view of the possible experimental realizability of our findings the model presented here should further be extended to include various dissipative processes that randomize the phase of coherent superposition states on very short time scales. Such an investigation would afford the inclusion of irreversible couplings on the basis of the master equation.

Non-classical states have great fundamental significance and are extremely appealing in their own right as a test of basic quantum theories as well as perhaps for practical applications. The idea of searching for non-classical states of phonons in solids is certainly expected to add a new dimension to the search for non-classical behaviour.

7. *Bibliography*

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